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Magnon-Mediated Indirect Exciton Condensation through Antiferromagnetic Insulators

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Electrons and holes residing on the opposing sides of an insulating barrier and experiencing an attractive Coulomb interaction can spontaneously form a coherent state known as an indirect exciton condensate. We study a trilayer system where the barrier is an antiferromagnetic insulator. The electrons and holes here additionally interact via interfacial coupling to the antiferromagnetic magnons. We show that by employing magnetically uncompensated interfaces, we can design the magnon-mediated interaction to be attractive or repulsive by varying the thickness of the antiferromagnetic insulator by a single atomic layer. We derive an analytical expression for the critical temperature $T_c$ of the indirect exciton condensation. Within our model, anisotropy is found to be crucial for achieving a finite $T_c$, which increases with the strength of the exchange interaction in the antiferromagnetic bulk. For realistic material parameters, we estimate $T_c$ to be around 7 K, the same order of magnitude as the current experimentally achievable exciton condensation where the attraction is solely due to the Coulomb interaction. The magnon-mediated interaction is expected to cooperate with the Coulomb interaction for condensation of indirect excitons, thereby providing a means to significantly increase the exciton condensation temperature range.

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Introduction.—Interactions between fermions result in exotic states of matter. Superconductivity is a prime example, where the negatively charged electrons can have an overall attractive coupling mediated by individual couplings to the vibrations, known as phonons, of the positively charged lattice. In addition to charge, the electron also has a spin degree of freedom. The electron spin can interact with localized magnetic moments through an exchange interaction exciting the magnetic moment by transfer of angular momentum. These excitations are quasiparticles known as magnons. Theoretical predictions of electron-magnon interactions have shown that these can also induce effects such as superconductivity [1–10].

Research interest in antiferromagnetic materials is surging [11,12]. This enthusiasm is due to the promising properties of antiferromagnets such as high resonance frequencies in the THz regime and a vanishing net magnetic moment. Much of this research focuses on interactions involving magnons or spin waves at magnetic interfaces in hybrid structures. Examples of this are spin pumping [13–19], spin transfer [15,20–22], and spin Hall magnetoresistance [23–28] at normal metal interfaces, and magnon-mediated superconductivity [9,10]. Recently, an experiment has also demonstrated spin transport in an antiferromagnetic insulator over distances up to 80 μm [29]. Moreover, antiferromagnetic materials are also of interest since it is believed that high-temperature superconductivity in cuprates is intricately linked to magnetic fluctuations near an antiferromagnetic Mott insulating phase [30,31]. Thus it is crucial to achieve a good understanding of antiferromagnetic magnon-electron interactions, as well as electron-electron interactions mediated by antiferromagnetic magnons.

In this Letter, we theoretically demonstrate the application of antiferromagnetic insulators to condensation of indirect excitons. An exciton is a bound state consisting of an electron and a hole. The excitons interact attractively through the Coulomb interaction due to their opposite charges [32]. Initially predicted many decades ago [33,34], the exciton condensate has been surprisingly elusive. A challenge is that the exciton lifetime is too short to form a condensate due to exciton-exciton annihilation processes such as Auger recombination [35–38]. The problem of short exciton lifetimes can be solved by having a spatial separation between the electrons and holes in a trilayer system, where the electrons and holes are separated by an insulating barrier [39–41] to drastically lower the recombination rate. Excitons in such systems are often referred to as (spatially) indirect excitons, and these are ideal to observe the exciton condensate. Herein, we consider a system where the insulating barrier is an antiferromagnetic insulator, as shown in Fig. 1. The insulating barrier can then serve a dual purpose: in addition to increasing the exciton lifetime, the spin fluctuations in the antiferromagnet mediate an additional attractive interaction between the electrons and the holes. This magnon-mediated interaction cooperates with
the Coulomb interaction thereby enabling an increase of the temperature range for observing exciton condensation in experiments. The exciton lifetimes achieved via antiferromagnetic insulators will be comparable to their nonmagnetic counterparts (~10 ns [42]), leaving the spin-independent physics unaltered.

The indirect exciton condensate has two main experimental signatures. The first is a dissipationless counterflow of electric currents in the two layers [43–45]. When the exciton condensate moves in one direction, the resulting charge currents in the individual layers are antiparallel due to the oppositely charged carriers in the two layers. The second signature is a large enhancement of the zero-bias tunneling conductance between the layers [46,47], reminiscent of the Josephson effect in superconductors. Comparing the critical condensation temperatures in trilayers with magnetic and nonmagnetic insulating barriers, that otherwise have similar properties and dimensions, should allow us to isolate the role of magnons in mediating the condensation.

The exciton condensate is expected to exist when the number of electrons in one layer equals the number of holes in the other. Thus far, to the best of our knowledge, experiments with an unequivocal detection of the exciton condensate have utilized quantum Hall systems with a half filling of the lowest Landau level to satisfy this criterion [48–52]. Such systems rely on high external magnetic fields. A recent experiment studying double-bilayer graphene systems has, however, been able to detect the enhanced zero-bias tunneling conductance signature of indirect exciton condensation without any magnetic field, by controlling the electron and hole populations through gate voltages [53]. This is an indication of the possible existence of an exciton condensate, and shows promise for finding a magnetic-field free exciton condensate.

In this Letter, we show that the magnon-mediated interaction between the electrons and holes can be attractive or repulsive depending on whether the two magnetic interfaces are with the same or opposite magnetic sublattices. In turn, this enables an unprecedented control over the interaction nature via the variation of the antiferromagnetic insulator thickness by a single atomic layer. Consequently, when the magnon-mediated interaction is paired with the Coulomb interaction, this can be used to control the favored spin structure of the excitons. In our model, we find that the critical temperature for condensation is enhanced by the exchange interaction in the antiferromagnet, and that a finite magnetic anisotropy is needed to have an attractive interlayer interaction around the Fermi level. Our results suggest that if one lets the insulating barrier in indirect exciton condensation experiments be an antiferromagnetic insulator, the magnon-mediated interactions can significantly strengthen the correlations between the electrons and holes.

Model.—We consider a trilayer system where an antiferromagnetic insulator is sandwiched between two fermon reservoirs, as illustrated in Fig. 1(a). We will then later consider the case where one of these reservoirs is populated by electrons, and the other by holes. This system can be described by the Hamiltonian \( \mathcal{H}_{\text{sys}} = \mathcal{H}_{\text{el}} + \mathcal{H}_{\text{mag}} + \mathcal{H}_{\text{int}} \), where \( \mathcal{H}_{\text{el}} \) describes the electronic part of the system in the fermon reservoirs, \( \mathcal{H}_{\text{mag}} \) describes the spins in the antiferromagnetic insulator, and \( \mathcal{H}_{\text{int}} \) describes the interfacial interaction between the fermions and magnons. We assume all three layers to be atomically thin, and thus two-dimensional, for simplicity.

We consider a uniaxial easy-axis antiferromagnetic insulator described by the Hamiltonian

\[
\mathcal{H}_{\text{mag}} = J \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j - K \sum_i S_i^2.
\]

Here \( J > 0 \) is the strength of the nearest-neighbor exchange interaction between the spins which have a magnitude \( |S_i| = \hbar S \) for all \( i \), and \( K > 0 \) is the easy-axis anisotropy constant. Next, we perform a Holstein-Primakoff transformation (HPT) [54] of the spin operators on each sublattice, denoted by sublattices \( A \) and \( B \), as defined in Fig. 1. From the HPT, we have that the operator \( a_i^{(\uparrow)} \) annihilates (creates) a magnon at \( r_i \) when \( r_i \in A \), and equivalently \( b_i^{(\uparrow)} \) annihilates (creates) a magnon at \( r_i \) when \( r_i \in B \). The magnetic Hamiltonian can be diagonalized through Fourier and Bogoliubov transformations to the form \( \mathcal{H}_{\text{mag}} = \sum_k \epsilon_k (\mu_k a_k + \nu_k a_k^\dagger) \). The magnon energy is given by \( \epsilon_k = \hbar \sqrt{(1 - y_k^2)\omega_k^2 + \omega_0^2(2\omega_k + \omega_0)} \), where \( k \) is the magnon momentum, \( y_k = z^{-1} \sum_\delta \exp (ik \cdot \delta) \), \( \delta \) a set of vectors to each nearest neighbor, \( z \) the number of nearest neighbors, \( \omega_k = \hbar JSz \), and \( \omega_0 = \hbar KS \). The eigenmagnon operators \( \mu_k^{(\uparrow)} \) and \( \nu_k^{(\uparrow)} \) are related to the HPT magnon

![FIG. 1. (a) An antiferromagnetic insulator (AFI) sandwiched between two separate fermion reservoirs, denoted by \( L \) and \( R \). We let the spins on sublattice \( A \) (illustrated in blue) be down, and the spins on sublattice \( B \) (illustrated in red) be up. (b) The fermions in the two reservoirs can interact through emission and absorption of magnons. For the process in the figure we have that either a spin-up fermion in \( L \) emits a \( S_z = +\hbar \) magnon (red arrow) which is absorbed by a spin-down fermion in \( R \), or a spin-down fermion in \( R \) emits a \( S_z = -\hbar \) magnon (blue dashed arrow) absorbed by a spin-up fermion in \( L \).](image)
operators through the Bogoliubov transformation $\mu_k = u_k a_k + v_k b_k^\dagger$, $\nu_k = u_k b_k + v_k a_k^\dagger$. The Bogoliubov coefficients $u_k$ and $v_k$ are given by $u_k = \sqrt{\Gamma_k + 1}/2$ and $v_k = \sqrt{\Gamma_k - 1}/2$, where we have introduced $\Gamma_k = \left(1 - |\omega_k|^2/(\omega_k + \omega_k^*) \right)^2)$. The effective coupling constants are defined so that they take on the value $J_k^L(r_l) = J_k^R$ if $r_l \in A_k^L$ and zero otherwise. We also have defined the electronic spin density

$$\hat{\rho}_j(r_i) = \frac{1}{2} \sum_{\sigma, \sigma'} \psi_{\sigma,j}(r_i) \sigma_{\sigma, \sigma'} \psi_{\sigma', j}(r_i).$$

(3)

with $\psi_{\sigma,j}^{(j)}$ annihilating (creating) a fermion with spin $\sigma$ in the $j$th ($j = L, R$) fermion reservoir, and $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ being a vector of Pauli matrices.

Effective magnon potential.—We will now use a path integral approach where we treat the magnon-fermion interaction as a perturbation, and integrate out the magnonic fields that give rise to processes as illustrated in Fig. 1. We compute that the coupling constants $J_{\mu, L/R}(q)$ describing the effective exchange coupling strength between the spin of the fermions in reservoirs $L, R$ to the spin of the eigenmagnons $\mu$, $\nu$ are $J_{\nu, L/R}(q) = v_{L, R}^* J_{\nu, L/R}^0(q - L/R)$ and $J_{\nu, L/R}^0(q) = v_{L, R}^* J_{\nu, L/R}^0(q - L/R)$. Since each interface is with only one sublattice, $J_{\nu, L/R}(q) = -u_{L, R} J_{\nu, L/R}^0(q)$ if the left interface is with sublattice $A$, and $J_{\nu, L/R}(q) = v_{L, R}^* J_{\nu, L/R}^0(q)$ if the left interface is with sublattice $B$. We get analogous results for the right interface. We see that the effective coupling constants $J_{\mu, L/R}(q)$ can have the same or opposite sign as the coupling constants $J_{\lambda, A,B}$ depending on which sublattice is at the interface. This has to do with the spin projection of the eigenmagnon relative to the equilibrium spin direction of the sublattice at the interface. The effective coupling constants $J_{\mu, L/R}(q)$ are also enhanced by a Bogoliubov coefficient $u_{L, R}$ or $v_{L, R}$ with respect to the coupling constants $J_{\lambda, A,B}$. These are typically large numbers. For $q = 0$ we have $u_{L, R} \approx v_{L, R} \approx 2^{-3/4} \times (\omega_{L, R}/\omega_{L, R})^{1/4}$ to lowest order in the small ratio $\omega_{L, R}/\omega_{L, R}$.
By studying Eq. (6), we note that we have $\text{Re}[U_\sigma(q, i\omega_n)] < 0$ for identical uncompensated interfaces, whereas for a system where one of the interfaces is with sublattice $A$ and the other with sublattice $B$, we have $\text{Re}[U_\sigma(q, i\omega_n)] > 0$. Consequently, this allows us to control whether the magnon-mediated interlayer-fermion potential $U_\sigma(q, i\omega_n)$ is attractive or repulsive by designing the interfaces. Whether this potential is attractive or repulsive can depend on a single atomic layer. This allows for an unprecedented high degree of control and tunability of the interlayer-fermion interactions. The sign difference of the potential can be explained by how the two fermions coupled by the magnon interact with the eigenmagnon spin. For $\text{Re}[U_\sigma(q, i\omega_n)] < 0$ we have processes where the fermions couple symmetrically to the magnon spin; i.e., both fermions couple either ferromagnetically or antiferromagnetically to its spin. On the other hand, for $\text{Re}[U_\sigma(q, i\omega_n)] > 0$ we have an asymmetric coupling, where one fermion couples ferromagnetically to the eigenmagnon spin and the other fermion couples antiferromagnetically.

**Indirect exciton condensation.**—We will now study spontaneous condensation of spatially indirect excitons where the attraction is mediated by the antiferromagnetic magnons. We consider the left (right) reservoir to be an n-doped (p-doped) semiconductor. We describe the semiconductors by the Hamiltonian

$$H_{\text{el}}(\tau) = \sum_{j=L,R} \sum_k \sum_{\sigma=\uparrow,\downarrow} e_j(k)\bar{\psi}_{\sigma,j}(k,\tau)\psi_{\sigma,j}(k,\tau),$$

with $e_j(k) = -e_F(k) = \hbar^2k^2/(2m) - e_F \equiv \epsilon(k)$. Here $m$ is the effective electron and hole mass, which we assume to be equal, and $e_F$ is the Fermi level. While the operator $\psi_{\sigma,L/R}^\dagger$ creates an electron with spin $\sigma$ in the left or right layer, we note that due to the negative dispersion in the right layer the excitations in this layer are effectively described by electron holes. We also note that we have not included a Coulomb interaction between the electron and the holes in our model. The effect of the Coulomb potential on indirect exciton condensation has been widely studied in previous literature [66]. We will later argue why the magnon-mediated potential is expected to cooperate with the Coulomb potential in the case of indirect exciton condensation.

The interaction in Eq. (5) is too complicated for us to solve for the exciton condensation. We then do an approximation similar to the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity [67,68], and assume that the dominant contribution to the interaction arises when the excitons have zero net momentum $(k + k' = q)$, and similarly for the Matsubara frequencies $(i\nu_j + i\nu_m = i\omega_n)$. Next, we introduce the order parameter

$$\Delta_\sigma(k, i\nu_m) \equiv -\sum_n \sum_{k'} U_\sigma(k - k', i\nu_m - i\nu_n) \times \psi_{\sigma,R}^\dagger(k', i\omega_n)\psi_{\sigma,L}(k', i\nu_n)$$

and its Hermitian conjugate, and perform a Hubbard-Stratonovich transformation of the effective action. By doing a saddle-point approximation and integrating over the fermionic fields [61], we then obtain the gap equation

$$\Delta_\sigma(k, i\nu_m) = \sum_m \sum_k \beta^{-1}U_\sigma(k - k', i\nu_m - i\nu_n) \times \frac{\Delta_\sigma(k, i\nu_m)}{\Delta_\sigma(k, i\nu_m)^2 + \epsilon(k)^2 + (\hbar\nu_m)^2}.$$  (9)

We note that the magnon-mediated potential is attractive when $U_\sigma(q, i\omega_n) > 0$ in the case of indirect exciton condensation, which can be seen from rearranging the fermionic fields in Eq. (5).

We now use Eq. (9) to find an analytical expression for the critical temperature $T_c$ below which the excitons spontaneously form a condensate. To obtain an analytical solution, we focus on the case when the gap functions and the magnon-mediated potential are independent of momentum and frequency. This corresponds to an instantaneous contact interaction, and we therefore assume that the gap functions have an s-wave pairing. Moreover, we see that the gap equation in Eq. (9) only has a solution when $\Delta_\sigma$ and $\Delta_\sigma$ have the same sign. In the case where spin degeneracy is unbroken, it is fair to assume that $\Delta_\sigma = \Delta_\sigma$, indicating tripletlike pairing. In superconductivity, s-wave and triplet pairing are mutually exclusive for even frequency order parameters, but in the case of indirect excitons the same symmetry restrictions on the order parameter do not apply, as the composite boson does not consist of identical particles. In other words, for indirect excitons the symmetries in momentum space and spin space are decoupled from one another. As both the magnon-mediated potential and the Coulomb potential are in the s-wave channel and the Coulomb potential is independent of spin, the magnon-mediated potential works together with the Coulomb potential enhancing the attractive exciton pairing interaction. The fact that we can design whether the magnon-mediated potential is attractive or repulsive allows us to control which spin channel is the most favorable for the excitons to condensate.

To determine $T_c$ we perform a BCS-like calculation [61,68] and restrict the sum over Matsubara frequencies to a thin shell around the Fermi level $(\hbar\nu_m < \epsilon_0)$, where the magnon-mediated potential is attractive. The analytical expression for $T_c$ is found to be

$$T_c = \frac{2e^{\gammaEM}\epsilon_0}{\pi k_B} \exp\left(-\frac{2\epsilon_0}{S\epsilon_0}\frac{2\epsilon_0}{\gamma_{\text{EM}}\gamma}\frac{J_{xL}J_{xR}}{\gamma}\right),$$  (10)

where $\gamma_{\text{EM}} \approx 0.577$ is the Euler-Mascheroni constant and $a$ the lattice constant of the semiconductors. Here we have assumed that the left and right magnetic interfaces consist of opposite sublattices. This leads to an attractive exciton interaction. If we assume the exchange energy among the spins in the bulk is much larger than the interface coupling.
optimal value, the critical temperature becomes squeezing is lost \[65\]. When the anisotropy takes on its moment of the magnon-mediated potential due to magnon

\[ \hbar \omega \equiv S ma^2 J^L_A J^R_B \] \[ \propto 10^{-13} \text{ s}^{-1} \] \[ [72] \]

and consequential so does the thin shell around the Fermi level where the magnon-mediated potential is attractive. In the case of a large anisotropy, \( \omega \gg \omega_{\text{opt}} \), the enhancement of the magnon-mediated potential due to magnon squeezing is lost \[65\]. When the anisotropy takes on its optimal value, the critical temperature becomes

\[ T_c^{\text{opt}} \equiv \frac{\sqrt{\hbar o_K S ma^2 J^L_A J^R_B}}{\sqrt{2 \pi} \epsilon^{1/2} k_B} e^{\epsilon M - 1/2} \]

Notably, we see that the critical temperature increases monotonically with increasing strength of the exchange interaction \( \hbar o_K \). The optimal choice of an antiferromagnetic insulator would then be a material with a magnetic anisotropy \( \hbar o_K \) on an energy scale proportional to the exchange coupling at the interface \( \hbar J^L_{A,B} \), and a very strong exchange interaction in the bulk of the antiferromagnetic insulator \( \hbar o_K \). As discussed in the Supplemental Material \[61\], inclusion of retardation and quasiparticle renormalization effects \[69–71\] via the Eliashberg method is expected to reduce the \( T_c \) estimated here by a factor between \( \sqrt{e} \) and \( e^{3/2} \). At the same time, accounting for the proper magnon dispersion leads to a similar increase \[70\] in \( T_c \) thereby leaving our estimate essentially unchanged after including these complications.

To show how high the \( T_c \) of indirect exciton condensation in our model can be using only the magnon-mediated interaction, we give a numerical estimate for realistic material parameters. Using the parameters \( S = 1, m \) equal the electron mass, \( a = 5 \text{ Å} \), \( \hbar J^L_A = \hbar J^R_B = 10 \text{ meV} \) \[7\], \( \epsilon_M = 8.6 \times 10^{-13} \text{ s}^{-1} \) \[72\], and assuming the magnetic anisotropy takes on its optimal value \( \omega_{\text{opt}} = 9.9 \times 10^6 \text{s}^{-1} \), we obtain a \( T_c^{\text{opt}} \) of approximately 7 K. Antiferromagnetic insulators that can be suitable for the proposed experiment are \( \text{Cr}_2\text{O}_3 \) \[63,73,74\] and NiO \[72\]. A possible emergence of a strong electric field across the barrier could in principle alter the magnetic properties in, e.g., \( \text{Cr}_2\text{O}_3 \) \[63,73\] and NiO \[74\]. We estimate the upper limit of a potential electric field to be around 47 V/mm, based on a “stress-test” scenario where 1% of the charge carriers have leaked through the insulating barrier, assuming a charge carrier density of \( 2.6 \times 10^{10} \text{ cm}^{-2} \) \[47\]. This estimate is considerably weaker than the requirements for influencing typical magnetic insulators \[63,73,74\]. In comparison to the critical temperature above, a recent experiment studying double bilayer graphene in the quantum Hall regime found the Coulomb-mediated exciton condensation to have an activation energy of \( \sim 8 \text{ K} \) \[52\], which was 10 times higher than what was found in an experiment using GaAs \[75\]. This demonstrates that the potential mediated by the antiferromagnetic magnons is capable of creating strong correlations between the electrons and holes that could significantly increase the critical temperature for condensation compared to when the excitons are just bound through the Coulomb interaction.

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